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Impacts of the 1900-1974 increase in anthropogenic aerosol emissions from North America and Europe on Eurasian summer climate

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ABSTRACT

9 The impact of North American and European (NAEU) anthropogenic
10 aerosol emissions on Eurasian summer climate during the twentieth century
11 is studied using historical single- and all-forcing (including anthropogenic
12 aerosols, greenhouse gases, and natural forcings) simulations from the Cou-
13 pled Model Intercomparison Project Phase 5 (CMIP5). Inter-model agree-
14 ment on significant linear trends during a period of increasing NAEU sulphate
15 emissions (1900-1974) reveals robust features of NAEU aerosol impact, sup-
16 ported by opposite changes during the subsequent period of decreasing emis-
17 sions. Regionally, these include a large-scale cooling and associated anticy-
18 clonic circulation as well as a narrowing of diurnal temperature range (DTR)
19 over Eurasian mid-latitudes. Remotely, NAEU aerosols induce a drying over
20 the western African and northern Indian monsoon regions, and a strengthen-
21 ing and southward shift of the sub-tropical jet consistent with the pattern of
22 temperature change. Over Europe, the temporal variations of observed tem-
23 perature, pressure, and DTR tend to agree better with simulations that include
24 aerosols. Throughout the twentieth century, aerosols are estimated to explain
25 more than a third of the simulated inter-decadal forced variability of European
26 near-surface temperature, and more than half between 1940 and 1970. These
27 results highlight the substantial aerosol impact on Eurasian climate, already
28 identifiable in the first half of the twentieth century. This may be relevant for
29 understanding future patterns of change related to further emission reductions.

30 **1. Introduction**

31 Increasing awareness and concern about the risks associated with the unfolding of climate
32 change, including changes in droughts and floods associated with shifts in precipitation patterns,
33 have posed considerable demand for credible climate projections at regional scale. Robust infor-
34 mation on regional impacts is crucial to be better prepared to manage and mitigate the impacts on
35 our society and the environment, including agriculture, energy, and water resources. Despite their
36 importance, reliable projections of regional climate change still remain a major challenge (IPCC,
37 2013).

38 Anthropogenic aerosols can drive changes in global and regional climate by impacting both
39 the energy and water cycles; yet, despite progress in the last decade, aerosols have remained the
40 dominant contributor to the uncertainty in total anthropogenic forcing over the industrial era in the
41 last three IPCC reports (IPCC, 2013). Uncertainties in quantifying the aerosol impact on climate
42 also hinder our ability to estimate the greenhouse gas (GHG) contribution to the recent global
43 temperature change (Hegerl et al., 1997; Stott and Jones, 2012; Ribes and Terray, 2013; Gillett
44 et al., 2013; Schurer et al., 2017). Although the IPCC (2013) assessed it as 'extremely likely'
45 that more than half of the observed warming since the mid-twentieth century was due to human
46 influences, the uncertainties in the separate effects of greenhouse gases and aerosols are much
47 larger. As emissions of GHGs and aerosols follow different pathways in the future, separation
48 of their respective effects is essential for improving confidence in climate projections for the 21st
49 century.

50 At global scale, aerosols are estimated to exert a net negative effective radiative forcing of -0.9
51 W/m^2 (-1.9 to -0.1 W/m^2 at 90% confidence level) on present-day climate, offsetting part of the
52 GHG-induced warming (Myhre et al., 2013). This negative radiative forcing results from pertur-

bation of the climate system through radiative and cloud micro-physical effects (aerosol-radiation and aerosol-cloud interactions, respectively; Boucher et al. (2013)), the relative magnitudes of which vary across aerosol species. In the direct effect (Charlson et al., 1992), aerosols scatter and absorb mainly solar radiation and thus modify the surface and atmospheric temperature distributions. The atmospheric heating thereby caused by strongly absorbing aerosols like black carbon can cause cloud changes by altering the surface energy budget and static stability as well as by cloud evaporation (burn-off); these changes are often referred to as the semi-direct effect (Hansen et al., 1997). Sulphate aerosols, on the other hand, scatter solar radiation, and are additionally efficient cloud condensation nuclei (CCN), which in turn affects cloud micro-physical properties and precipitation processes – the cloud albedo (first indirect) (Twomey, 1977) and the cloud lifetime (second indirect) effect (Albrecht, 1989). Non-linearities in these indirect aerosol effects, caused by saturation effects in the cloud-aerosol interactions, can cause aerosols to be more important in a more pristine atmosphere (e.g. Carslaw et al. (2013)).

Being emitted mainly by fossil fuel and biomass burning sources, present-day global atmospheric levels of sulphate, organic carbon, and black carbon are substantially larger than those at pre-industrial times. Historical changes, however, have neither been linear nor spatially homogeneous. They result mainly from a combination of changes over two macroregions: Emissions from North America and Europe (NAEU) increased steadily from pre-industrial times to the peak in the 1970s, when air quality legislation led to their reduction. Asian emissions, in contrast, started to rapidly increase in the 1950s as a result of rapid economic development and have been increasing ever since (Lamarque et al., 2010; Hoesly et al., 2017). These geographical differences in the emission time series, combined with the short lifetime of aerosols in the atmosphere, result in spatially and temporally heterogeneous distributions and higher concentrations around the source regions. The global mean impact of aerosols is therefore not necessarily representative of regional climate

77 responses, which can be substantial (e.g. Ramanathan et al. (2001)) even when the magnitude of
78 global forcing remains relatively small.

79 Numerous studies have suggested that anthropogenic aerosols had an important impact on the
80 twentieth century climate over various regions (Boucher et al., 2013; Bindoff et al., 2013). Emis-
81 sions of aerosols from human activities predominantly located in the northern hemisphere (NH)
82 have for instance been linked to a southward shift of the Inter-Tropical Convergence Zone since the
83 early 1900s by muting the warming of the northern hemisphere relative to the southern hemisphere
84 (Chiang and Friedman, 2012; Rotstayn and Lohmann, 2002; Hwang et al., 2013). Aerosols have
85 been shown to have played a predominant role in the decrease of northern hemisphere monsoon
86 precipitation (Polson et al., 2014), as well in weakening its regional components over East Asia (Li
87 et al., 2016; Guo et al., 2013), South Asia (Bollasina et al., 2013; Undorf et al., 2018), and North
88 West Australia (Rotstayn et al., 2012). Furthermore, aerosols were found to have contributed to the
89 decrease of precipitation in West Africa (Ackerley et al., 2011; Dong et al., 2014), to cause tropical
90 contraction (Allen and Ajoku, 2016), and to have modulated the Atlantic multi-decadal circulation
91 and North Atlantic storms (Booth et al., 2012; Dunstone et al., 2013). Aerosol reductions in recent
92 decades might have amplified Arctic temperature increase and sea-ice loss (Acosta Navarro et al.,
93 2016). Yet, many of these findings are associated with large uncertainties and are, at times, contro-
94 versial, resulting in vivid discussions across the scientific community. IPCC (2013) noted that the
95 "lack of agreement across studies prevents generalization of findings to project aerosol-induced
96 changes in regional atmospheric circulation or precipitation in the near term".

97 This is also true for studies focused on European climate, where changes in surface tempera-
98 ture and diurnal temperature range (DTR) have been related to aerosol-induced shift in the surface
99 solar radiation regime from dimming to brightening (Wild et al., 2007; Makowski et al., 2008,
100 2009). DTR is a measure of high-frequency temperature variability and is not only important for

101 climate impacts as diverse as crop yields (Lobell et al., 2007) and human mortality and morbidity
102 (Kim et al., 2016), but has also been suggested to be a useful indicator to separate aerosol and
103 GHG forcing (Schnur and Hasselmann, 2005; Wild et al., 2007): Through reducing surface solar
104 radiation, aerosols are expected to reduce maximum (Tmax) more than minimum (Tmin) temper-
105 ature, causing a decrease in DTR, while GHG interact with outgoing long-wave radiation and are
106 thus expected to impact Tmax and Tmin equally (although a preferred response of TMIN to GHG
107 warming has also been suggested (Rohde et al., 2013a)). Other studies found the recent reduction
108 in aerosols to have increased the frequency of light precipitation events despite no changes in total
109 precipitation in observations (Stjern et al., 2011) and an increase in the ratio between convective
110 to stratiform precipitation in a GCM (Stjern and Kristjánsson, 2015). Complex interactions un-
111 derpin also the relationship between European aerosol loading and the North Atlantic Oscillation
112 (Chiacchio et al., 2011; Pausata et al., 2014).

113 Located upstream of the large Eurasian landmass and just north of Africa, European aerosols
114 in particular have thus potential impact on huge inhabited areas, both through local impacts and
115 through remote impacts mediated by atmospheric circulation adjustments. Their impact through-
116 out the twentieth century, however, has not been consistently investigated so far in a multi-model
117 framework, particularly for the earlier part of the century when aerosol effects might have been dis-
118 proportionately large in a cleaner atmosphere (Carslaw et al., 2013). This is a gap this study aims
119 to address. We make use of experiments carried out as part of the Coupled Model Intercomparison
120 Project Phase 5 (CMIP5) initiative (Taylor et al., 2012). These comprise of multiple realizations of
121 experiments of all historical forcings as well as individual forcings with a range of coupled global
122 climate models, which allows us to analyze the effect of anthropogenic aerosol and compare it to
123 those of other forcing factors, both when acting in isolation and when combined. The availability
124 of a large set of simulations allows the identification of robust patterns of forced climate response

125 by (1) averaging over multiple members, thus reducing the effect of internal variability, and (2)
126 sampling across various model formulations.

127 In order to provide a basic characterization of the climate response to NAEU aerosol emissions
128 expected from CMIP5 simulations, we focus on the boreal summer season, when the aerosol im-
129 pact on temperature and other variables mediated by temperature differences, such as the position
130 of the ITCZ and the jet streams, is expected to be largest due to the insolation maximum in the
131 northern hemisphere (see discussion in Hegerl et al. (1997)). The analysis examines changes in
132 near-surface temperature (TAS), precipitation, diurnal temperature range (DTR), sea level pres-
133 sure (SLP), and zonal wind at 300 hPa. This subset, although limited, allows us to identify the
134 aerosol impact on relevant surface climate features, some of which are long-term observed, and
135 investigate the potential of aerosols to bring about changes in the atmospheric circulation.

136 The remainder of the manuscript is organized as follows: After a short description of the datasets
137 and the analysis methods used (Section 2), we illustrate the spatio-temporal changes in anthro-
138 pogenic aerosol emissions and the resulting atmospheric aerosol loading and radiative effect (Sec-
139 tion 3). The linear trends in the all-forcing and single-forcing CMIP5 ensembles during the histor-
140 ical period of increasing NAEU sulphate emissions are then shown, contrasted by those during the
141 later period of decreasing NAEU emissions (Section 4). While this captures multi-decadal to near-
142 centennial changes, higher-frequency variability of European near-surface temperature, sea level
143 pressure, and DTR are analyzed by means of area-mean time series also from observations, and
144 the contribution of each single-forcing factor to the total simulated forced inter-decadal variability
145 is estimated (Section 5). The study is completed by summary and conclusions (Section 6).

2. Data and methods

We use data from the CMIP5 twentieth century historical experiments, and select models which provide simulations forced only with anthropogenic aerosols (Tab. 1). In addition to this ensemble of aerosol-only simulations ("AA"), we also analyze simulations with forcing from well-mixed greenhouse gases only ("GHG"), natural forcings only (solar radiation and volcanoes, "NAT"), and all these forcings combined ("ALLF") for the same set of models. Note that ALLF are the standard historical simulations, which are expected to reproduce the observations. For each model, we choose ensembles of GHG, NAT, and ALLF with the same size as the ensemble available for AA, so that the results from the different ensembles are comparable. Results from the trend analysis are shown for the multi-model mean ("MMM"), obtained by first averaging the individual ensemble members for each model and successively averaging these ensemble means. Given the different ensemble sizes -ranging from one to five- this ensures that each model is given equal weight.

While the chosen subset has a more comprehensive representation of aerosol effects compared to the full suite of CMIP5 models (Taylor et al., 2012), there is still substantial variation amongst the models (Ekman, 2014): In GFDL-CM3 and NorESM1-M, aerosol micro-physics and chemistry including cloud droplet number concentration (CDNC; first indirect effect) are simulated online, and precipitation formation depends on this CDNC (second indirect effect). CSIRO-Mk3.6.0, HadGEM2-ES, and CanESM2 also have online aerosols schemes, but use diagnostic CDNC formulations, on which precipitation does (CSIRO-Mk3.6.0, HadGEM2-ES) or does not (CanESM2) depend. GISS-E2-R and IPSL-CM5A-LR prescribe aerosol fields from offline calculations, use diagnostic CDNC representations, which precipitation does (GISS-E2-R) or does not

168 (IPSL-CM5A-LR) depend on. On top of these differences, the chemistry models used and/or the
169 way of the diagnostic derivation of CDNC may vary as well.

170 Given that all models are forced with the same aerosol emission inventories (Lamarque et al.,
171 2010), the use of multiple models does thus not only reduce the contribution from internal vari-
172 ability, but allows us moreover the identification of features which are robust to these differences
173 in the models' representation of indirect aerosol effects as well as their aerosol distribution and
174 background (pre-industrial) aerosol concentrations (Wilcox et al., 2015). This is important since
175 these inter-model differences have been found to cause a large spread in the aerosol responses
176 (Boucher et al., 2013; Kasoar et al., 2016)). Robustness is assessed by comparing the sign of the
177 response in the MMM with that in the models' ensemble means.

178 The models considered span the full range of CMIP5 spread in climate sensitivity (Flato et al.,
179 2013), and represent the diversity in the effective radiative forcing due to direct and indirect aerosol
180 effects combined (Zelinka et al., 2014). Their historical all-forcing MMM reproduces the clima-
181 tology and observed changes in surface temperature patterns, and simulates the climatology of
182 many large-scale circulation features reasonably well (Flato et al., 2013; Kumar et al., 2016). For
183 DTR, too, the all-forcing simulations from chosen CMIP5 models separately as well as the MMM
184 of all models' all-forcing simulations agree well with observations over Europe both in terms of
185 climatology (Lindvall and Svensson, 2014; Cattiaux et al., 2015; Liu et al., 2016) and long-term
186 trends (Lewis and Karoly, 2013).

187 The aerosol forcing in all models is based on decadal aerosol emissions with an annual cycle.
188 We compute boreal summer (June, July, and August, "JJA") means from monthly-mean data, and
189 all model data is re-gridded prior to further analysis to match the model with the lowest resolution,
190 which is $3.75^{\circ} \times 2.8^{\circ}$. For the comparison with observations, we instead interpolate the model
191 data to the observational resolution and mask according to the observational coverage. Anomalies

192 from the climatological state are considered, and the climatologies are computed from the models’
193 pre-industrial control simulations.

194 Simulated temperature and sea-level pressure changes are compared to those derived from avail-
195 able observational data. For near-surface temperature anomalies, we use the HadCRUT dataset
196 version 4.5 (“HadCRUT”) (Morice et al., 2012), the CRUTS dataset Version 3.24.01 (“CRU”)
197 (Harris et al., 2014), the gridded Berkeley Earth Surface Temperature dataset (“BEST”) (Rohde
198 et al., 2013b), and GISS Surface Temperature Analysis (“GISTEMP”) (GISTEMP Team, 2017;
199 Hansen et al., 2010). For DTR, we use CRU and BEST, and for sea level pressure we use the Met
200 Office Hadley Centre mean sea level pressure data set HadSLP2 (1850-2004, “HadSLP”) (Allan
201 and Ansell, 2006).

202 We compute DTR from the CMIP5 model data as the difference between monthly-mean Tmax
203 and monthly-mean Tmin, as provided by the modelling groups (see Tab. 1), and follow the same
204 procedure for the BEST dataset; for the CRU data, monthly-mean DTR is used as provided by
205 Harris et al. (2014). Note that the observational datasets differ in their station input sources, time
206 sampling, quality control, homogenization, and area-averaging; for a comprehensive analysis of
207 the relevance of these differences see Thorne et al. (2016).

208 Long-term changes in climate are investigated by computing linear trends, calculated as least-
209 squares regressions, and displayed as change per decade. The robustness of the simulated trends
210 is measured by the agreement among the various models: Stippled areas are regions where at
211 least all but one of the models’ ensemble means (i.e. 5 out of 6 or 6 out of 7, depending on the
212 availability of some variables for specific experiments) agree on sign. The time period common to
213 all model simulations is 1860-2005, but we limit the analysis to 1900 onward when observations
214 are less sparse (Morice et al., 2012). Trends in the NAT ensemble are not shown for brevity
215 unless otherwise stated, as they are found to be small compared to those in the AA and GHG

ensembles as well as barely anywhere robust across models for the variables, time periods, and regions considered in this analysis.

In order to support the use of linear trends to identify the link between aerosol loading and near-surface temperature, a joint empirical orthogonal functions (EOF) analysis is also performed (Deser and Blackmon, 1993; Wang et al., 2016). This method, which is an ordinary EOF analysis of the two variable fields combined in space, decomposes the data into orthogonal modes each consisting of one pattern per variable and a common time series, ordered by the fraction of combined variance explained. In contrast to a regression analysis, this identifies covarying patterns without the need to presume timeseries or spatial patterns. Prior to the EOF analysis, the data are temporally smoothed in order to suppress inter-annual variability. This is done by taking subsequent 11- and 7-year running means which improves filter characteristics relative to taking running means only with a single window length (e.g. von Storch and Zwiers (1999)). For a sensitivity analysis see supplementary Text S1.

3. Spatio-temporal changes in aerosol emissions and associated near-surface temperature

Sulphur dioxide (SO₂) is the main component of anthropogenic aerosol emissions (Lamarque et al., 2010; Hoesly et al., 2017). The time series of total SO₂ emissions from various regions illustrate the dominance of NAEU emissions from pre-industrial times until the mid-twentieth century and show the peak in the 1970s, with the trend reversing in Europe around 1975, following a slightly earlier reversal in the US (Fig. 1). While Asian (incl. eastern Russian) emissions also started to increase before 1970, they do not exhibit strong (Japan, Russia) or any (China, India, etc.) downward trends afterwards. This is also visible in the spatial pattern of the linear trends of SO₂ emissions during 1900-1970 and 1971-2010 (Fig. 2).

238 The corresponding trend patterns in column-integrated sulphate content (sulphate loading) and
239 aerosol optical depth at 550 nm (AOD) in the CMIP5 model ensemble show that aerosols spread
240 further east of the source region towards central Eurasia as a result of transport by the climato-
241 logical westerlies (Fig. 2). Although CMIP5 models use the same aerosol emission inventories,
242 the simulated aerosol distribution patterns may vary from model to model due to differences in
243 meteorology, chemical parameterizations, and pre-industrial background concentrations (Carslaw
244 et al., 2013; Wilcox et al., 2015; Kasoar et al., 2016). Nonetheless, there is considerable agree-
245 ment on the sign of the trends over Eurasia (Fig. 2c-d). The emissions of other aerosol species
246 (black carbon, organic carbon) changed over time less homogeneously in space (Figs. S1-4), but
247 the simulated changes in AOD, which integrates the amount and optical properties of all simulated
248 aerosol species, are very similar to those in sulphate loading, thus reflecting the dominance of
249 sulphate aerosols (Fig. 2e-f).

250 A joint EOF analysis reveals modes of covariability of sulphate loading and near-surface tem-
251 perature (TAS) as simulated in the AA ensemble for summer over the NH excluding the Pacific
252 (Fig. 3). The dominant temperature signature is a large-scale cooling associated with an overall
253 increase in sulphate aerosols until they peak in the 1970s (Fig. 3a). The first mode's patterns and
254 time series for both sulphate loading and TAS are very similar to those of the first mode from
255 EOFs of the variables separately (Fig. S5a-b), which confirms that these patterns represent a sub-
256 stantial part of variability in both sulphate loading and TAS individually in the CMIP5 ensemble.
257 Separate EOF analyses of other variables show that approximating the aerosol-related impact with
258 linear trends captures a large fraction of their variability in the CMIP5 AA ensemble (for example
259 precipitation and DTR, Fig. S5c-d).

260 The higher modes from the joint EOF analysis reflect differences in the time series of aerosol
261 emissions and associated temperature impacts at smaller spatial scales. The sulphate loading and

temperature patterns of the second mode (Fig. 3b), for instance, has opposite signs in North America and Europe compared to Asia. The principal components accordingly resemble the time series of the difference in sulphate loading over these regions. However, since only a very small fraction of the covariance is explained by the higher modes, we focus here on the dominant behavior as reflected in the first mode.

4. Simultaneous long-term changes in NAEU emissions and near-surface climate

Given the strong opposite trends in SO₂ emissions from NAEU before and after the peak in the 1970s, it is reasonable to expect their climate signature to also show a trend reversal around the same time. Emissions from elsewhere, e.g. Asia, in contrast, started to increase in the 1950s and might thus have perturbed the climate during the first period, but we expect their continued increase during the second period to result in changes of the same sign in both periods, albeit of different magnitudes. This also holds true for GHG forcing, which has increased during both periods. We thus identify changes during 1900-1974 in the ALLF ensemble as dominantly driven by NAEU aerosol forcing if they are robust and have the same sign as those in the AA but different to those in the GHG or NAT ensembles. This approach is further validated by contrasting the trends with those during 1975-2005, when we expect the decreasing NAEU emissions to drive changes of the opposite sign. Note, however, that the impact of emissions from, say, Asia is not negligible during 1975-2005, so that the effect of decreasing NAEU emissions might be offset by Asian emissions.

Despite clear limitations, this approach allows us to disentangle the NAEU aerosol imprint in presence of other forcings (provided they changed monotonously during the whole record). The advantage over other methods (e.g., EOF analysis) is that it allows us to assess inter-model consistency and to compare with the other ensembles and observations, for which aerosol data are not available.

Based on the timing of the trend reversal in simulated sulphate loading (indicated by a black line in the time series in Fig. 3), we choose 1900-1974 and 1975-2005 as the time periods with increasing and decreasing aerosols, respectively. Note that choosing instead the partition year 1970, as done for the trends in emissions because they are decadal only, does not affect the results reported below (not shown). The first time period includes the slow increase in emissions (Fig. 1), so that trends per decade are smaller during the first than the second period; European area-mean (35-60°N, 0-60°E) trends in the aerosol-only MMM amount to +2.0 and -2.6 mg/m²/decade for sulphate loading and +0.028 and -0.046 1/decade for AOD during 1900-1975 and 1975-2005, respectively.

Because emissions from both North America and Europe reversed sign around this time, we will in the following attribute the changes to NAEU emissions. The larger trends in European emissions and their closer proximity to the area studied, however, might suggest them to have a larger share of the impacts at least on Eurasian near-surface temperature.

a. Near-surface temperature & sea level pressure

The CMIP5 AA ensemble shows a widespread decrease in near-surface temperature over the Atlantic-Eurasian region during the period of increasing NAEU emissions (Fig. 4a). This trend is reversed in the later period with a warming of similar magnitude as the earlier cooling over central Europe, and a slightly weaker warming over the Asian mid-latitudes (Fig. 4b). Note that over eastern China, temperatures decrease during both periods consistent with trends in local aerosol emissions (Fig. 2a,c), so NAEU emissions are not the (sole) driver of the cooling over this region in the CMIP5 models. The larger trends during the second period -shown in Fig. 4 scaled by a factor of one half- compared to those during the first period are consistent with the larger trends in emissions, sulphate loading, and AOD (Fig. 2).

GHG forcing produces warming during both periods (Fig. 4e-f), and both the AA and the GHG signature are robust across the models in the whole region. In the ALLF ensemble, which has the two anthropogenic forcings counteract each other in the first period, the models agree on the mid-latitude cooling as well as the warming at higher and lower latitudes. The zonally extended temperature anomalies are consistent with Shindell et al. (2010), who found the influence of inhomogeneous radiative forcing to extend three to four times further in the zonal than in the meridional direction. The vertical cross-section of temperature trends averaged over 30-60°N shows moreover that although the strongest cooling is located near the surface over Europe, the temperature signal extends up to the mid- and upper-troposphere and as far eastward as central Asia (Fig. S6a-b), suggestive of an eastward propagation from Europe. Natural forcing does not result in appreciable trends in the CMIP5 models (not shown).

Sea level pressure, consistent across the models, shows anomalous anticyclonic trends over the Eurasian mid-latitudes in the first period both in the AA and the ALLF ensemble, which is consistent with the atmospheric adjustment to the surface cooling (Figs. 4a,c; S7a,c). The largest aerosol-induced anomalies in TAS and SLP east of the area of highest emission changes (Figs. 4, S6-7) is consistent with expectations from aerosol transport as well as temperature advection by climatological westerlies (not shown). While increasing anthropogenic aerosols thus dominate the simulated Eurasian mid-latitude temperature changes during 1900-1974, simulated tropical temperature changes appear to be more strongly influenced by GHG forcing, with a widespread warming seen both in the GHG and the ALLF CMIP5 ensembles (Fig. 4c-f).

b. Jet stream strength and position

The zonal character of the aerosol-driven temperature changes as simulated with the CMIP5 models suggests that aerosols might also affect circulation features in the latitudinal direction

331 via changes in the meridional temperature gradients. We find indeed that both the AA and the
 332 ALLF ensemble show a strengthening of the equatorward side of the subtropical jet over Asia,
 333 as identified from changes in 300-hPa zonal wind strength, during the first time period, and a
 334 weakening during the second period (Fig. 5a-d). GHG (and NAT, not shown), on the other hand,
 335 show no large-scale significant or robust trends (Figs. 4e-f; 5e-f). This is consistent with the
 336 thermal wind relation, with AA cooling the mid-latitudes more than the tropics and thus enhancing
 337 the temperature gradient, which results in a strengthened zonal wind (Figs. 4a,c; 5a,c). Conversely,
 338 during the second period, the decrease in NAEU emissions warms the mid-latitudes more than
 339 the tropics, thus reducing the gradient and weakening the zonal wind (Figs. 4b,d; 5b,d). The
 340 significant positive wind speed anomalies on the southern flank of the climatological jet position
 341 during the first period, and negative ones during the second, can be additionally interpreted as a
 342 southward and northward, respectively, displacement of the circulation in response to the changing
 343 meridional temperature gradients.

344 *c. Precipitation*

345 Another feature associated with the change in the meridional circulation is the shift of the inter-
 346 tropical convergence zone (ITCZ) in response to variations in the inter-hemispheric temperature
 347 gradient, driven by the aerosols' preferential cooling of the Northern vs. the Southern hemisphere
 348 (Chiang and Friedman, 2012; Hwang et al., 2013). In the CMIP5 models, we also see an aerosol
 349 signal on precipitation over western Africa, where rainfall is largely controlled by the ITCZ (Un-
 350 dorf et al., 2018): A drying during the period of increasing NAEU aerosols, and a wettening during
 351 the period of decreasing aerosols, both in the AA and the ALLF ensemble (Fig. 6a-d).

352 Over most of Eurasia, where climatological precipitation is relatively low and spatially hetero-
 353 geneous, precipitation trends in the CMIP5 AA ensemble during 1900-1974 are small (Fig. 6a).

354 Key robust features are a wettening trend over western Europe as well as a drying over the high-
355 latitudes, including Scandinavia and Northern Russia, which is also reversed during 1975-2005
356 (Fig. 6b). The ALLF ensemble shows also that if aerosols are included, the drying over Europe as
357 simulated for GHG forcing only is restricted to the Mediterranean (Fig. 6c-f).

358 In the subtropical and tropical monsoon regions South Asia, East Asia, western Africa, in con-
359 trast, we see a robust aerosol signal during both periods (Fig. 6a-d). During the first period, the
360 AA ensemble shows a drying over northern India, which reverses during the second period (Fig.
361 6a-b). This robust and significant aerosol signal also dominates -although over a smaller domain-
362 in the ALLF ensemble, despite the counteracting effect of GHGs (Fig. 6c-f).

363 Both the AA and the ALLF ensemble also show a meridional tripole pattern of longitudinally-
364 elongated bands featuring a drying over central China and the western Pacific, a wettening over
365 southern China and Indochina, and again a drying over the Maritime Continent during the first pe-
366 riod (Fig. 6a,c). This pattern is especially consistent between AA and ALLF over land, whereas the
367 GHG impact appears to emerge in driving the oceanic precipitation increase over the Indian Ocean
368 and the western Pacific (Fig. 6e). While these precipitation changes have thus been clearly driven
369 by anthropogenic aerosols in the CMIP5 models, our results do not allow us to unequivocally sep-
370 arate the role of remote and local aerosols, owing to the simplicity of the linear-trend approach: In
371 addition to NAEU emissions, local emissions also increased -again potentially more effective than
372 later in the century-, the effects of which presumingly added onto those from NAEU emissions
373 (Guo et al., 2013). The importance of NAEU aerosols is clear, however, from comparison with a
374 shorter, earlier period (1900-1950) for which the assumption of negligible Asian emissions change
375 is even more justified (Fig. 1): The precipitation changes during 1900-1975 when both NAEU and
376 Asian emissions increased strongly resemble those during 1900-1950, when only NAEU emis-
377 sions increased, so that the response to NAEU forcing must be a substantial part of the response

378 to the combined forcing (Figs. 6, S10). Impact from NAEU aerosols in our results is plausible,
 379 too, since the anomalous anticyclone over mid-latitude Asia, resulting from the large-scale near-
 380 surface and tropospheric cooling (Figs. 4, S6-7) might cause northeasterly winds which oppose
 381 the climatological monsoon flow and thus induce a weakening of the East Asian summer monsoon
 382 circulation, as visible in the precipitation changes over East Asia (Figs. 6). During 1975-2005, on
 383 the other hand, the impact of increasing local emissions could have counteracted that of decreasing
 384 NAEU emissions (Guo et al., 2013), and the continued drying suggests that NAEU emissions were
 385 not the most important factor in driving the precipitation changes over China and South-East Asia
 386 in the CMIP5 simulations during this later period (Fig. 6a-b).

387 *d. Diurnal temperature range*

388 DTR shows a widespread decrease over much of the European and Asian mid-latitudes both in
 389 the AA and the ALLF ensemble during the first time period (Fig. 7a,c). Over Europe, the trend
 390 is reversed during the second period (Fig. 7b,d). The trends in the GHG ensemble (and NAT, not
 391 shown) are less uniform and not consistent with ALLF (Fig. 7e-f), which suggests that aerosols,
 392 not greenhouse gases, dominate the twentieth century DTR variations over Europe, in agreement
 393 with expectations. Interestingly, a significant, consistent aerosol signal is also noticeable at low
 394 latitudes (from northern Africa to India), but of opposite sign to that in the mid-latitudes, with
 395 GHG (and NAT) trends again much smaller, less robust, and inconsistent with those in the ALLF
 396 ensemble; coinciding precipitation trends (Fig. 6) hint at modulations in diurnal convection by
 397 changes in cloudiness as a possible mechanism underlying this aerosol control on DTR over these
 398 regions, which is supported by trends in total cloud fraction (Fig. S8).

399 *e. Sensitivity study and early twentieth century*

400 The robustness of the trends during 1900-1974 is further ascertained by considering the shorter
401 1900-1950 period, during which Asian emissions increased even less and were at levels compara-
402 ble to those of combined European and North American emissions in 1850 (Fig. 1). During this
403 period, trends of similar patterns to those for 1900-1974 emerge in the CMIP5 simulations over the
404 region considered, but with smaller magnitudes, as expected from the smaller increase in aerosols
405 (and GHGs) which had taken place by then (Figs. S9-11). For near-surface temperature, the cool-
406 ing in the AA ensemble during 1900-1950 does not dominate the ALLF response, but reduces the
407 warming trends over Europe seen in the GHG ensemble in the mid-latitude band which shows
408 aerosol-related cooling during 1900-1974. The pattern of change in precipitation over South-East
409 Asia described above is already recognizable in the AA trends during 1900-1950; also, DTR shows
410 a robust narrowing over the mid-latitudes in both the AA and the ALLF ensemble.

411 These findings suggest that aerosols were already a key driver of regional climate anomalies
412 before the peak aerosol increase between 1950-1970s, for some variables even dominant over
413 GHGs. Note that saturation effects might contribute to this by potentially making the aerosol
414 emissions less effective later in the twentieth century (e.g. Carslaw et al. (2013)). The similarity
415 of the patterns and their already considerable magnitude during 1900-1950 furthermore support
416 the notion that remote (NAEU) emissions contributed substantially to the simulated near-surface
417 climate variation over Russia and Asia, since the amount of remote aerosol emissions dominated
418 over that of local emissions even more in 1950 than in 1970: In 1950 (1970), there were 8 (4)
419 times more emissions from Europe than from Asia, and from 1900-1950 (1900-1970), those from
420 Europe increased nearly 6 (3) times faster than those from Asia (Fig. 1).

421 5. Temporal evolution of European climate

422 *a. Comparison with observations*

423 In this section, the model-based findings on long-term changes associated with aerosols dis-
424 cussed above are complemented by an analysis of area-mean time series for the region with the
425 largest trends in the AA and ALLF ensembles (35° - 60° N, 0° - 60° E) (Fig. 8). We focus on near-
426 surface temperature, sea-level pressure, and DTR, which all show a spatially homogeneous re-
427 sponse across this region in the CMIP5 ensemble (Fig. 4,7). These time series are also compared
428 with observations, the coverage of which is better for this region than most others especially early
429 in the twentieth century.

430 The temporal change in near-surface temperature shows the contrast between the warming in
431 the GHG-only experiments and the cooling in the AA ensemble (Fig. 8a). The small gradients
432 in both ensembles in the first decades of the twentieth century is consistent with weak variations
433 in anthropogenic emissions, while clear differences between the gradients in the two ensembles
434 start appearing around 1925, with similar magnitudes of opposite sign until the 1970s. The largest
435 aerosol-induced variations are found during 1940-1975, followed by a plateau and a weak recov-
436 ery, reflecting the peak and subsequent decline in global aerosol emissions. In contrast, the GHG
437 warming increased exponentially during the twentieth century, with the largest trends after 1960.
438 The all-forcing ensemble reflects the modulation due to both drivers as well as the contribution
439 from natural variability, for example the volcanic cooling (e.g. after the Agung eruption in 1963,
440 e.g. Robock and Mao (1995)). After a few decades of negligible anthropogenic forcing at the be-
441 ginning of the century, temperature variations are dominated by aerosol cooling until at least 1970,
442 when GHG warming starts to dominate instead. Comparison with the observed changes shows that
443 the CMIP5 models are able to capture the observed twentieth century variations of near-surface

temperature over Europe (observations within the 95% range of ALLF). The most notable feature in the observations is the cooling during 1940-1980, which is consistent with the prevailing role of anthropogenic aerosols.

For sea level pressure (Fig. 8b) and DTR (Fig. 8c), the single-forcing ensembles diverge less clearly, reflecting higher variability, lesser model agreement, and more complex responses of these variables compared to temperature to the different forcings. For SLP, both the AA and the ALLF ensemble encompass the observations, but the GHG and NAT ensembles do not, indicating that aerosol forcing is essential to explain the observed variations.

For DTR, the two observed datasets agree on multi-decadal variability, showing a decrease until 1930, followed by a steep increase, and a subsequent decrease between the mid-1930s and the mid-1980s. They differ in their absolute anomalies, however, due to discrepancies in the first half of the century (Thorne et al., 2016). While all ensembles are compatible with the CRU observations at the 95% level, the GHG and NAT ensembles are incompatible with the BEST dataset in the second half of the twentieth century. The AA and ALLF ensembles are the only ones to show a DTR narrowing over most of the twentieth century as do the observations. This overall provides further confirmation of aerosol forcing being necessary to explain historical observed variations. However, due to strong influence of internal climate variability, conclusive attribution of observed changes was not possible.

b. Contribution of individual forcings to simulated European summer climate

The contribution of each single forcing on the forced component of simulated European inter-decadal climate variability can be estimated by calculating the gradient of the multi-model mean time series from each ensemble and comparing the magnitude of those from the single-forcing CMIP5 ensembles (AA, GHG, NAT) to their arithmetic sum as in Wilcox et al. (2013). This

467 approach assumes that the role of internal variability is negligible in the multi-model mean and
468 that the responses to the individual forcings add linearly (Fig. 9). The time series are smoothed
469 prior to the analysis by consecutively taking 11- and 7-year running means to suppress variability
470 on time scales shorter than inter-decadal (Section 2).

471 For European near-surface temperature, the sum of the gradients (AA+GHG+NAT) is a rea-
472 sonable approximation to the gradient from the ALLF ensemble (Fig. 9a, top), and during the
473 twentieth century, the contribution of AA to the total forced simulated variability (36%) is com-
474 parable to that of GHG (43%), and larger than that of NAT (21%). The aerosol impact is even
475 dominant during 1940-1970, with AA explaining more than 50% of the total forced variability.
476 The aerosol contribution to the total forced inter-decadal model variability in sea level pressure is
477 also discernible and amounts to about a third throughout the twentieth century, and slightly more
478 again in the period 1940-1970 (Fig. 9b). For DTR, AA contributes on average a forth throughout
479 the twentieth century; this contribution grows towards the later decades, but then the sum of the
480 gradients of the single-forcing time series approximates the all-forcing one less well (Fig. 9c).

481 **6. Summary, discussion, and conclusions**

482 The long-term signature of anthropogenic aerosol emissions mainly from North America and
483 Europe (NAEU) on Eurasian summer climate throughout the twentieth century has been identified
484 in an ensemble of coupled climate (CMIP5) models. The analysis was motivated by the need to
485 advance current understanding of the effects of aerosols on regional climate, which is of utmost
486 importance to more confidently assess and quantify the drivers of past climate variations as well
487 as to reduce uncertainties in near-future climate projections. North America and Europe were
488 the key aerosol emission regions worldwide for most of the twentieth century, with the potential to
489 influence climate downstream over large inhabited regions. Yet, the topic has not been consistently

490 investigated so far: While earlier studies have mostly focused either on individual variables or used
491 only one climate model, robust information on the aerosol impact on a range of relevant variables
492 across multiple coupled models is still largely unknown.

493 The aerosol imprint on simulated summer climate was inferred from inter-model agreement on
494 linear trends during a period of increasing NAEU sulphate emissions (1900-1974), contrasted with
495 trends during the more recent period of decreasing emissions (1975-2005). This was supported
496 by an analysis of the covariability of near-surface temperature and sulphate loading. The main
497 findings are:

- 498 • Regionally and during the 1900-1974 period, aerosols generated a large-scale anomalous
499 cooling stretching from Europe across most of the Eurasian mid-latitudes in the CMIP5 mod-
500 els. The cooling, largest at the surface, extends to the mid and upper troposphere, and is
501 associated with a large-scale anticyclonic sea-level pressure anomaly centered over Russia,
502 as well as with a widespread narrowing of DTR. Aerosols also strengthened the northern
503 hemisphere subtropical jet on its equatorward side, which is consistent with changes in the
504 meridional temperature gradient, and decreased monsoon precipitation over western Africa,
505 northern India, and eastern China.
- 506 • During the 1975-2005 period, most of these changes are reversed, which provides support
507 to their association with NAEU sulphate aerosol emissions – only precipitation changes over
508 East Asia, where strong positive trends in local aerosol emissions are expected to superpose
509 the effect of decreasing NAEU emissions, do not reverse.
- 510 • Analysis of observations averaged across Europe shows clear evidence for aerosols and GHGs
511 both being important for models to reproduce inter- and multi-decadal variations in near-
512 surface temperature. Observed variations in sea-level pressure are noisy, but tend to agree

much better with simulations that include anthropogenic aerosols than with those without aerosols. Observed DTR has very high data uncertainty, but shows a decline over much of the century that does not occur in the CMIP5 GHG and NAT ensembles.

- Assuming linearity among the single forcing responses, anthropogenic aerosols are estimated to explain on average more than a third of the forced simulated inter-decadal variability in European near-surface temperature during the twentieth century, and more than half during 1940-1970.

The time-series analysis thus highlights the importance of aerosols in addition to GHGs for explaining temporal variations in observed European near-surface climate, in agreement with Bindoff et al. (2013). For DTR, our results add onto those by Makowski et al. (2008), providing the evidence from an ensemble of CMIP5 experiments. Furthermore, we show that regionally AA explain a substantial fraction of simulated European climate inter-decadal variability from as early on as 1900, which is even larger than that found by Wilcox et al. (2013) for global mean temperature.

Significant changes in strength and latitudinal position of the subtropical jet stream during the twentieth century have been discussed in a number of studies, including those focusing on variations of the tropical belt width, given that the jet stream can be interpreted as the poleward boundary of the tropics. While some studies only looked at annual means (Archer and Caldeira, 2008b; Fu and Lin, 2011; Moore, 2013) or the winter season (Strong and Davis, 2007), changes in winter and summer, if any, were generally found to be consistent in sign (Archer and Caldeira, 2008a; Hudson, 2012; Davis and Birner, 2013; Pena-Ortiz et al., 2013; Abish et al., 2015), despite the pronounced climatological seasonality. Evidence suggests a weakening and/or poleward shift of the NH subtropical jet since 1979 from satellite observations (Fu and Lin, 2011), reanalyses (Archer and Caldeira, 2008a; Hudson, 2012), and observations (Davis and Birner, 2013; Pena-Ortiz et al.,

2013). Together with weaker and/or even opposite trends since the mid-twentieth century (Strong and Davis, 2007; Archer and Caldeira, 2008b; Pena-Ortiz et al., 2013; Abish et al., 2015) and ice core proxy data (Moore, 2013), this indicates a strengthening before 1979. In the framework of tropical expansion/contraction, these changes have been previously linked to sulphate forcing (Allen et al., 2012, 2014; Allen and Ajoku, 2016; Ming et al., 2011), especially during the summer (Tao et al., 2016), but the sign reversal associated with the evolution of aerosol forcing throughout the 20th century has not been previously discussed in the context of jet stream changes, and the appearance of noticeable trends as early as the beginning of the century has not been shown yet.

The remote impact on monsoon precipitation found here in the CMIP5 models is consistent with Polson et al. (2014). For western Africa, observations show similarly a drying during 1950-1985, and a recovery thereafter (Nicholson, 2013), and the region has been identified as sensitive to aerosol forcing (e.g. Rotstayn and Lohmann (2002); Ackerley et al. (2011)). Dong et al. (2014), who found both Asian and European sulphur dioxide emissions relevant for the decreased West African precipitation, and Undorf et al. (2018), who attributed observed precipitation changes over West Africa during 1920-2005 mainly to NAEU emissions, both used a single model. Our results, based on a range of CMIP5 models, also indicate the predominant importance of NAEU emissions on summer monsoon precipitation over western Africa. The overall weakening of the South Asian monsoon during the second half of the twentieth century forced by anthropogenic aerosols has been acknowledged before (e.g. Bollasina et al. (2011)). While other studies found regional and remote aerosols responsible in varying ratios (e.g. Bollasina et al. (2014); Guo et al. (2015)), we show that considering the periods with increasing and decreasing remote emissions separately indicates remote emissions to be most important. For East Asia, we found remote emissions not dominant during 1975-2005, consistent with strong impact of local aerosols (e.g. Guo et al. (2013)), but our results do not exclude substantial impact of remote aerosols, especially

560 before 1975, and are not inconsistent with a mechanism of remote aerosol impact on monsoon
561 precipitation found in earlier studies (Cowan and Cai, 2011; Dong et al., 2015).

562 The models used in this study explore the range of climate sensitivity and aerosol forcing of
563 those CMIP5 models that include a representation of indirect effects, from those with weak (e.g.
564 IPSL-CM5A-LR) to those with strong (e.g. GFDL-CM3) indirect effects (Section 2). Previous
565 studies, however, showed substantial differences between the response to historical aerosol emis-
566 sions in CMIP5 models with and without a representation of aerosol indirect effects (Wilcox et al.,
567 2013; Guo et al., 2015); other models, which neglect indirect aerosol effects, might therefore
568 misrepresent the response to aerosol emissions as identified in this study.

569 While the use of historical single- and all-forcing simulations allows us to identify features of
570 aerosol impact within the system of interactions and feedbacks -insofar represented in the models-,
571 targeted simulations would allow a more rigorous disentanglement of the effects of aerosols from
572 different source regions, including NA and EU. Similarly, the CMIP5 models' differing aerosol
573 representations increase the robustness of our findings, but also prevent us from exploring the
574 mechanisms of aerosol impact further. A future study with a different focus would thus com-
575 plement our work and help understand better the physical mechanisms mediating NAEU aerosol
576 impact.

577 To conclude, the findings add valuable information about the robustness of the aerosol impacts
578 as well as new insights on their climate footprint, such as their modulation of DTR and the jet
579 stream. The symmetry of the simulated impacts during the period of increasing and of decreas-
580 ing emissions also sheds light on the changes to be expected from future reductions in aerosol
581 emissions from NAEU and elsewhere.

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919 **LIST OF TABLES**

920 **Table 1.** CMIP5 models used in this study. The Table includes the number of ensemble
921 members available for aerosol-only simulations as well as whether each model
922 includes any representation of the first and second indirect aerosol effects. An
923 overview of the experiment design is given in Taylor et al. (2012). 44

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925 for aerosol-only simulations as well as whether each model includes any representation of the first and second
926 indirect aerosol effects. An overview of the experiment design is given in Taylor et al. (2012).

Institute ID	Model Name	#ens. members	aerosol indirect effects		Reference
			1st	2nd	
CCCMA	CanESM2	5	x	-	von Salzen et al. (2013)
CSIRO-QCCCE	CSIRO-Mk3.6.0	5	x	x	Jeffrey et al. (2013)
IPSL	IPSL-CM5A-LR	1	x	-	Dufresne et al. (2013)
MOHC	HadGEM2-ES	4	x	x	Collins et al. (2011)
NASA GISS	GISS-E2-R	5	x	x	Schmidt et al. (2014)
NCC	NorESM1-M	1	x	x	Iversen et al. (2013)
NOAA GFDL	GFDL-CM3	3	x	x	Donner et al. (2011)

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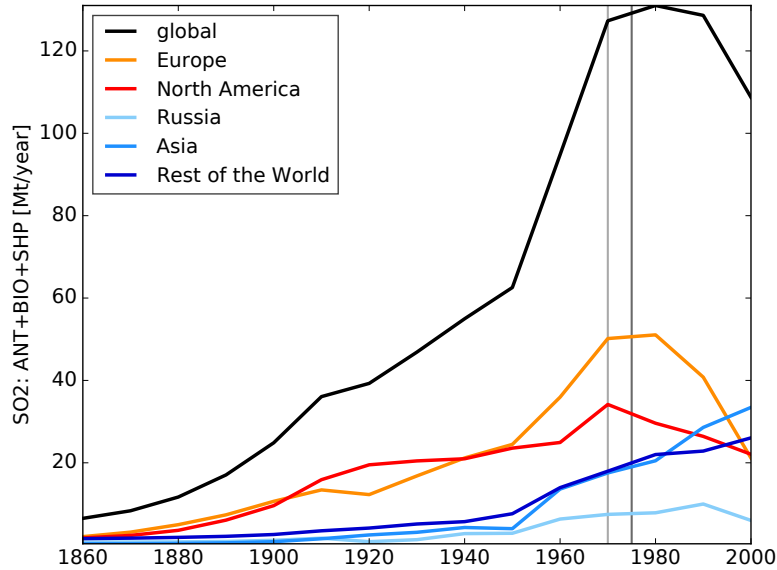


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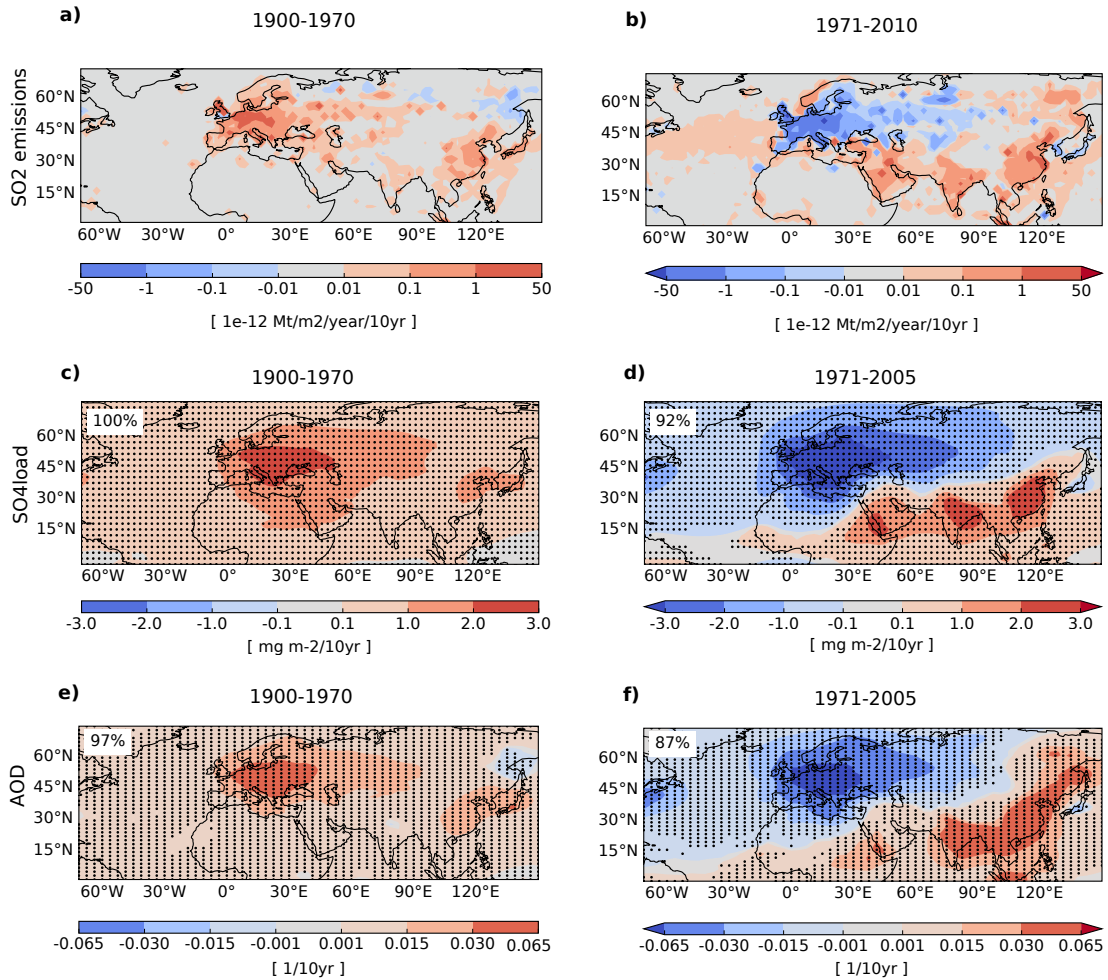


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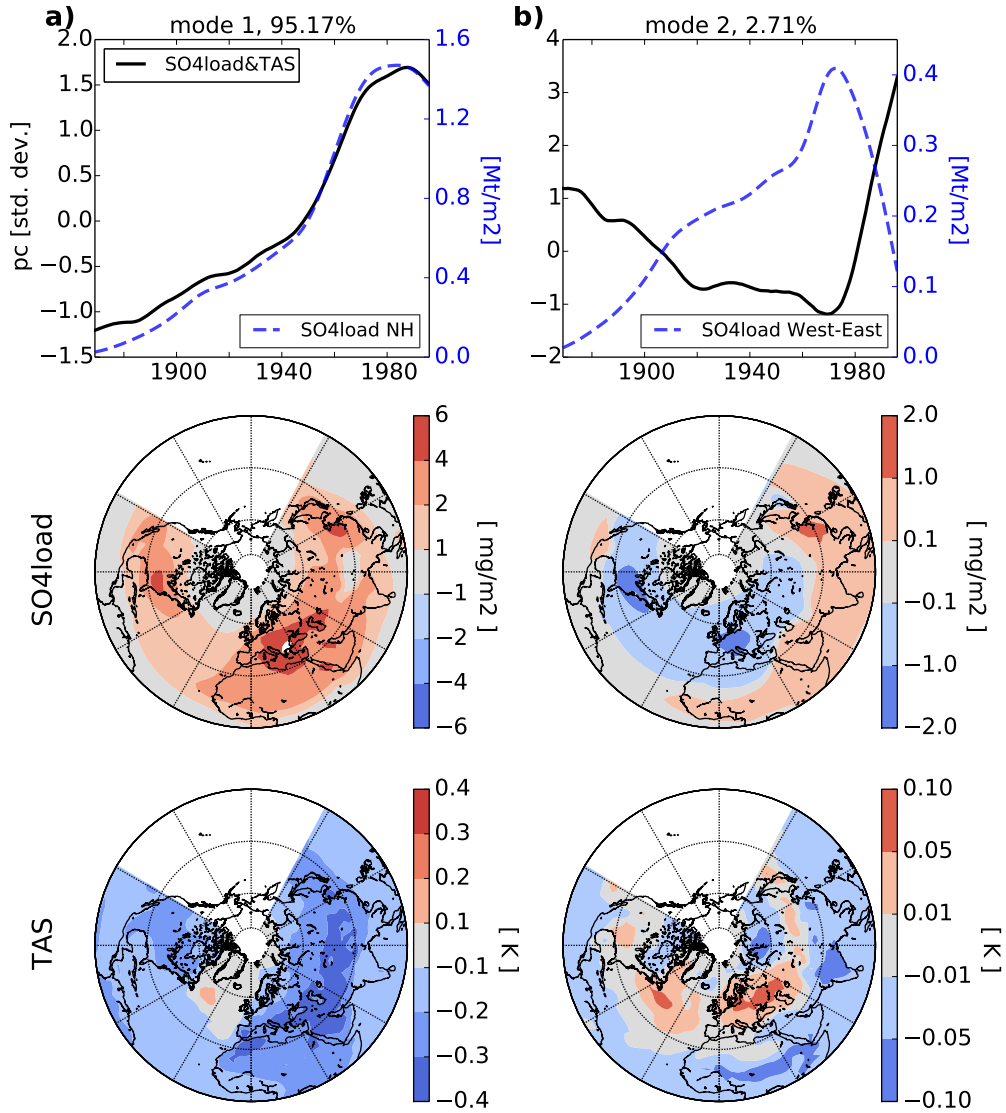


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1900-1974 TAS trends

1975-2005 TAS trends (x0.5)

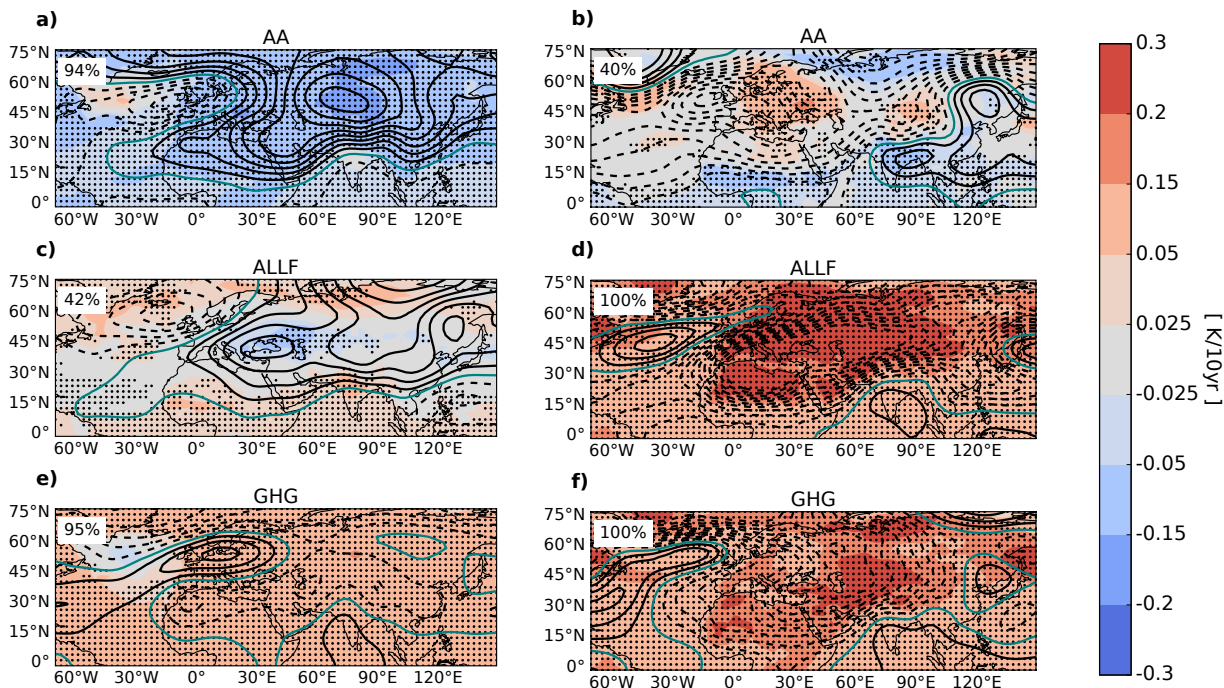


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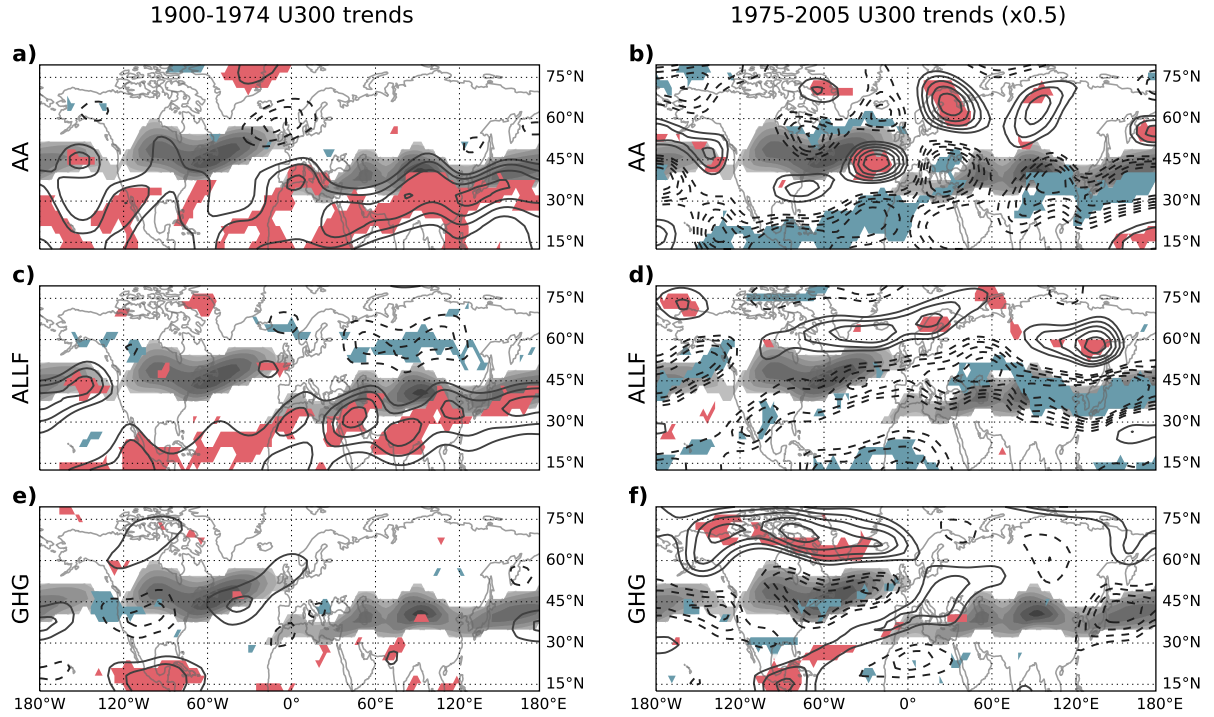


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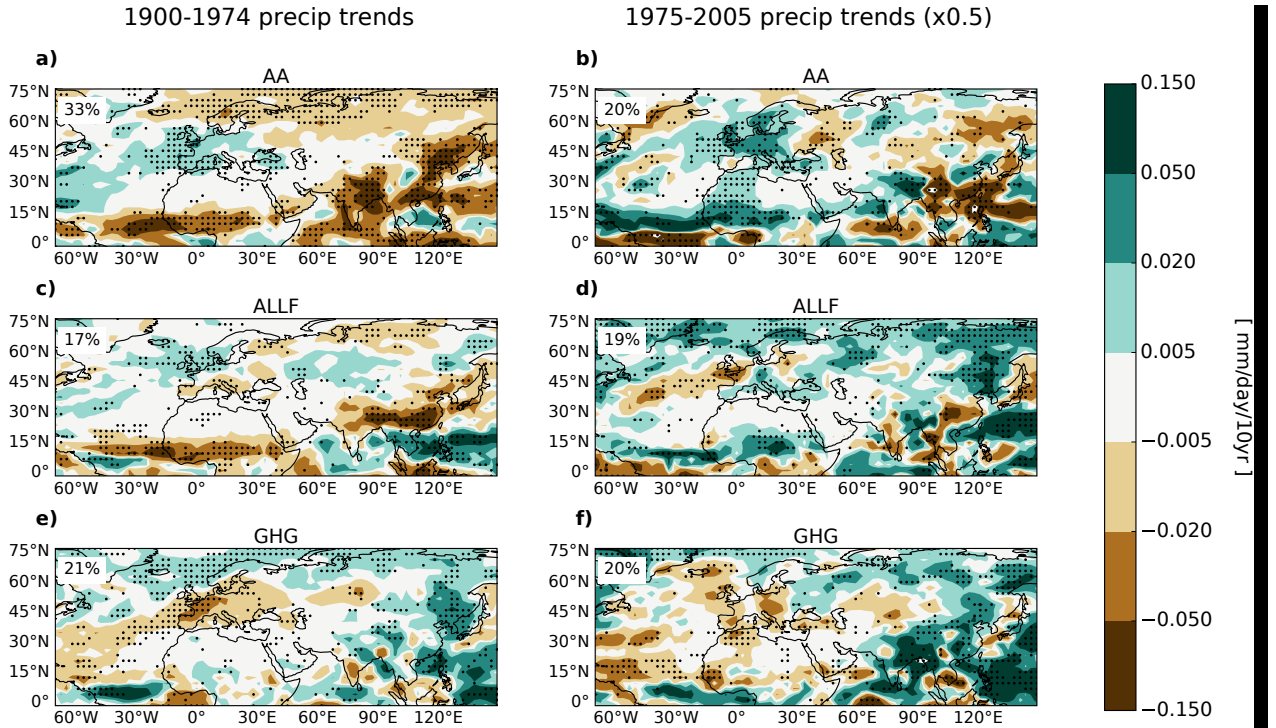


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1900-1974 DTR trends

1975-2005 DTR trends (x0.5)

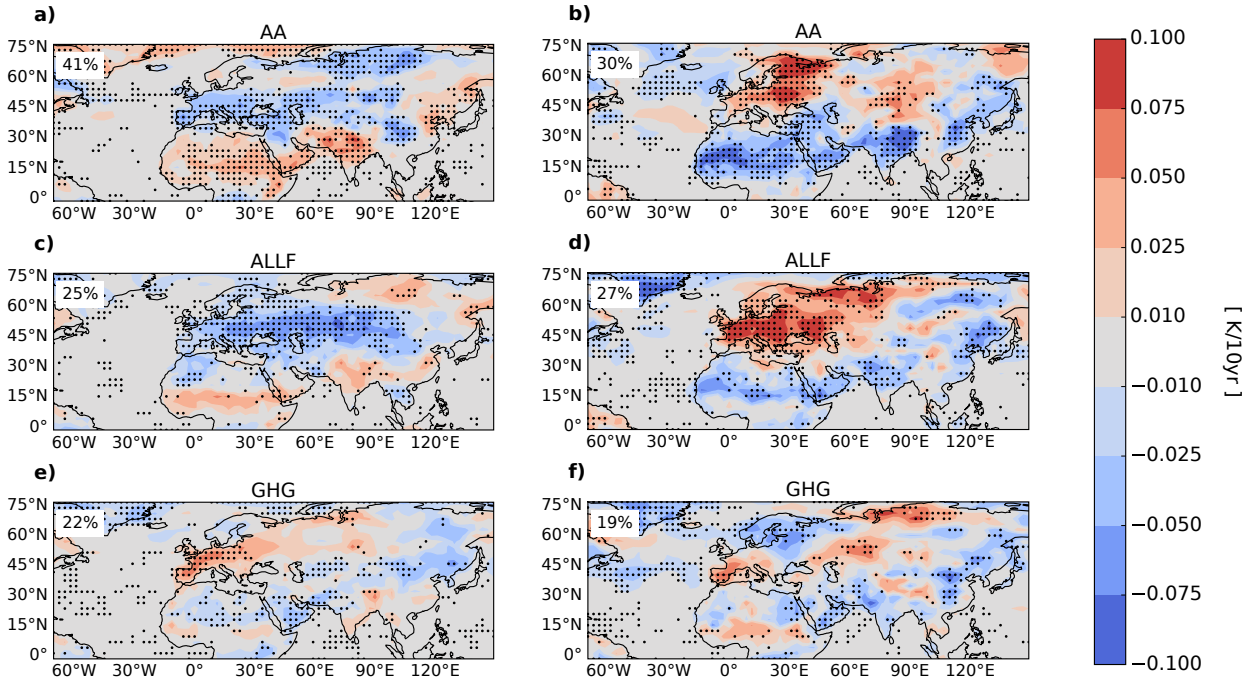


FIG. 7. Linear trends in DTR (in K/decade) for JJA during (left) 1900-1974 and (right) 1975-2005 for the multi-model mean of the (top) AA, (middle) ALLF, and (bottom) GHG simulations for the models listed in Tab. 1; for AA, no HadGEM2-ES data is available, thus the MMM of the remaining six models is shown. Stippling indicates where at least 6 out of the 7 (ALLF, GHG) or 5 out of the 6 (AA) models' ensemble means agree on sign, and the fraction of stippled grid points within 0-90°N, -30-120°E is given in the top left corner. Note that trends during the second period are scaled by 1/2 for easier comparison with those during the first period.

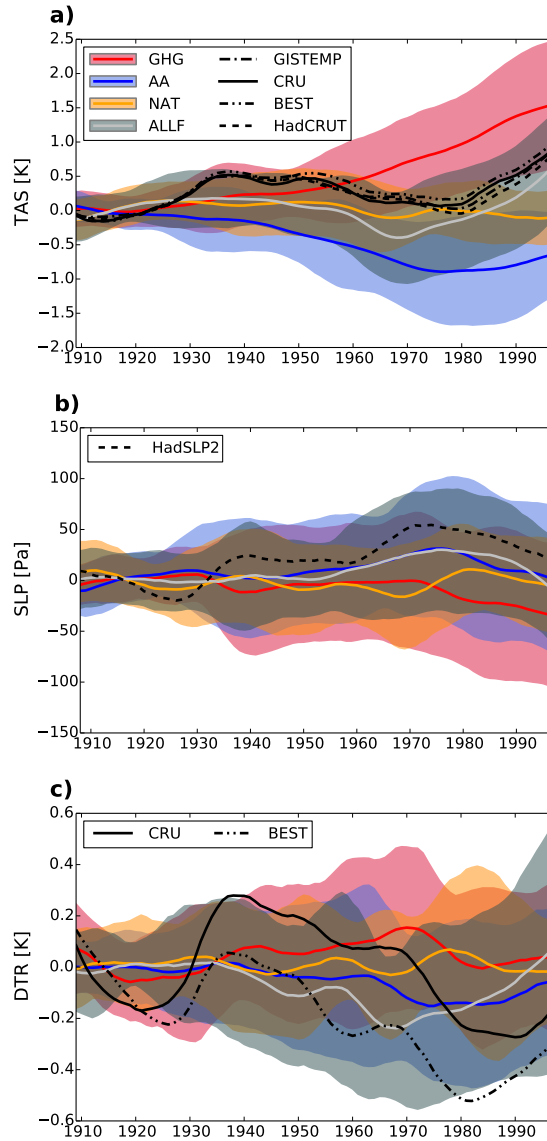


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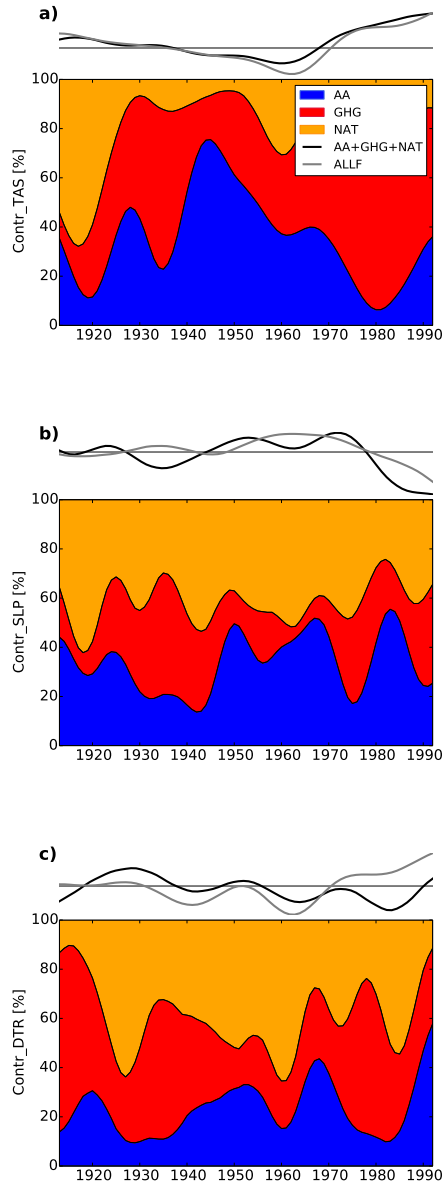


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